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Corrosion Models in Non-Isothermal LBE Coolant Loops

Corrosion of containment and structural materials presents a critical challenge in the use of liquid lead-bismuth eutectic (LBE) or lead as a nuclear coolant in accelerator-driven systems (ADS) and advanced reactors. The majority of previous corrosion research focuses on the influence of the local conditions. However, corrosion and precipitation rates and distributions can depend strongly on global conditions such as the global temperature profile and global system geometry, limiting the applicability of many corrosion models. Precise simulations of all hydrodynamic conditions encountered in practice by use of flowing conditions in the laboratory are difficult and expensive if not entirely impossible. Nevertheless, it is important to understand the flow-induced corrosion mechanism in order to control and reduce the corrosion processes in such systems subject to corrosion. Therefore, theoretical analyses are very necessary to provide general information on the flow-induced corrosion. Theoretical results can be used to explain and to apply the limited experimental results on LBE corrosion and provide useful information on the design and operation of LBE coolant systems.

I. Improved Application of Local Models to Steel Corrosion in LBE Loops

Several corrosion models have been developed in aqueous media based on the experimental results. Assuming the bulk concentration to be zero, Balbaud-Celerier and Barbier [*Journal of Nuclear Material*, 289, 277 (2001)] applied these models to calculate corrosion rates in liquid-metal loops. The models predicted much higher values than the experimental data. The number and quality of the data are not sufficient to differentiate the scaling behavior of the corrosion rate based on these models. Therefore, an accurate description of the corrosion phenomena that take place in a non-isothermal system can be accomplished only if the axial conditions are taken into consideration**.** To find out the bulk concentration effects, we improve the previous local models [J. Zhang and N. Li, *Nuclear Technology*, 144 (12), (2003)] when applied to closed loops by incorporating some global condition effects. In particular, the bulk corrosion product concentration is calculated based on balancing the dissolution and precipitation in the entire closed loop. Mass transfer expressions developed in aqueous medium and an analytical expression are tested in the liquid-metal environments. The improved model is applied to a pure lead loop and produces results closer to the experimental data than the previous local models do. The model is also applied to a lead-bismuth eutectic (LBE) test loop. Systematic studies illustrate the effects of the flow rate, the oxygen concentration in LBE and the temperature profile on the corrosion rate. The results showed that the corrosion rate in LBE closed loops could be reduced by carefully controlling the oxygen concentration in LBE.

II. Kinetic Corrosion Model for Simple Non-Isothermal LBE Loops

Regarding surface reactions, a kinetic corrosion model for a simple loop with constant cross-section was developed through solving the mass transfer governing equation based on some reasonable assumptions [J. Zhang and N. Li, *Journal of Nuclear Materials*, 321, 184 (2003)]. Comparing with previous local corrosion models, the present kinetic model considers the effects of the temperature profile. The non-local analyses provide the corrosion and precipitation distributions in the entire LBE loop and determine the value of the highest corrosion and precipitation and where they occur. The model was applied to calculate the corrosion/precipitation rates and distributions for different operate conditions in the DELTA loop, a test loop set up at LANL to study the corrosion of various materials under an LBE environment. It was shown that the highest corrosion/precipitation does not necessary occurs at the highest/lowest temperature. The locations depend on the temperature profile. For the DELTA loop, we found the highest corrosion occurs at the beginning of the test leg, while the highest precipitation occurs shortly after the test leg where the temperature decreases significantly. Comparisons between the results of loop flow and those of the open pipe flow with the same flow and temperature conditions illustrate the different behaviors of the corrosion/precipitation in open pipe flow and closed loop flow.

Employing the present kinetic corrosion model, the effects of the maximal temperature and the temperature difference between the maximal and minimal temperature are studied. For the DELTA loop, simulation results indicate that the corrosion rate increases with the temperature difference. The change slows after the difference exceeds 100ºC, and nearly saturates after reaching 200ºC. For the same gradient, the corrosion increases drastically with the maximal temperature. For accelerated corrosion testing, it is desirable to increase the testing temperature and set the temperature difference closer to 200ºC. This new understanding of the dependence of corrosion/precipitation rates on the axial temperature profile is useful for helping design and operate non-isothermal closed loop systems. We plan to verify the key aspects in future experiments. This dependence implies that the corrosion test results obtained from one flow loop cannot be directly applied to another loop with a different temperature profile. It also suggests that it is possible to design flow systems to minimize corrosion and precipitation or to change the locations of maximal precipitation for enhanced system lifetime performance.

We also examined the variations of the corrosion product concentration in the bulk flow along the loop axis in the steady state [J. Zhang and N. Li, *Journal of Nuclear Science and Technology*, submitted 2003]. It was found that the iron bulk concentration was not zero. It varies along the axis and reaches the highest value at the end of the test leg (the highest temperature leg). The variation becomes smaller with the increasing bulk velocity. However, even for a small bulk velocity, the variation is small enough to be neglected. For example, the difference between the maximum and the minimum bulk concentrations for $V = 0.1$ m/s is less than 3% of the average bulk concentration. Therefore, it is reasonable to assume the concentration of corrosion product in the bulk flow to be constant along the axis at the steady state for the DELTA loop.

III. A Correlation for Steel Corrosion in Non-Isothermal LBE Loops

Analyzing the corrosion rate variations in the DELTA loop, a correlation between the average corrosion rate and hydraulic conditions at the hottest leg in a non-isothermal LBE loop was developed [J. Zhang and N. Li, *Journal of Nuclear Science and Technology*, submitted 2003]. The average Sherwood number in the highest temperature leg (test section) is correlated to the loop conditions by the dimensionless equation:

$$\mathrm{Sh}_{av} = 0.53\,\mathrm{Re}^{0.6}\,\mathrm{Sc}^{1/3}(\mathrm{d/L})^{1/3}(\Delta\mathrm{T}/\mathrm{T}_{max})^{1/3},$$

where Re and Sc are the Reynolds and Schmidt numbers, respectively, d(m) is the pipe diameter, L(m) is the loop length, ΔT (ºC) is the temperature difference between the highest and lowest temperature, and T_{max} (ºC) is the maximal temperature. The correlation indicates that the average corrosion rate at the highest isothermal leg in a non-isothermal liquid-metal loop is proportional to $(\Delta T/T_{max}L)^{1/3}$ as determined by the axial loop conditions.

Analyses on the corrosion/precipitation profiles in non-isothermal LBE loops indicated that if the corrosion/precipitation rate scales $Re^{0.6}Sc^{1/3}(d/L)^{1/3}D/d$, the non-dimensional corrosion/precipitation rate is independent of the hydraulic factors for high Reynolds numbers in the diffusion limited regime. The corrosion/precipitation profile is determined solely by the surface temperature profile or the resultant surface corrosion product concentration profile.

IV. Transient Corrosion/Precipitation Behaviors in Non-Isothermal LBE Loop

The kinetic model for simple non-isothermal LBE loops was extended to unsteady-state cases [J. Zhang and N. Li, *Corrosion*, accepted 2003]. An analytical solution is obtained by solving the mass transfer equation in the boundary layer for diffusion-limited corrosion processes. The temperature-dependent wall corrosion product concentration is a function of the stream-wise coordinate. Solutions for different wall concentration profiles are used to examine the transient process. The initial and the final behaviors of the corrosion/precipitation profile are shown for different loop flows. These results reveal important differences between the initial and steady-state corrosion/precipitation phenomena and how quickly they evolve. This new understanding will help improve the interpretation of the experimental data and the rational application of the corrosion test data to experimental and industrial systems.

The transient corrosion/precipitation distribution depends on both the local and global conditions. Corrosion occurs everywhere in the loop at the beginning and precipitation appears at some positions in later times. At the steady state, the highest corrosion/precipitation does not occur at the location with the highest/lowest temperature (or surface corrosion product concentration); the corrosion product concentration in the bulk flow is almost uniform and the average value equals to the average value of the surface concentration for a simple loop. The axial diffusion contribution can be neglected for high Reynolds numbers, and the corresponding corrosion/precipitation distribution profile is independent of the flow velocity and determined by the temperature (surface concentration) profile. The transient term related to the global concentration profile plays an important role at the early times, while the transient term related to the mean surface concentration dominates the transient process at later times.

For the DELTA loop, the average corrosion rate at the maximal temperature section (the primary test section) decreases rapidly in time and reaches the steady-state value after about 200 s. This time is nearly independent of the temperature gradient and the oxygen concentration in LBE. The decrease is more significant further downstream, calling attention to the need to record the test locations and properly interpret the test results. The corrosion rate increases with the increasing temperature gradient and the decreasing oxygen concentration. This model provides location-resolved corrosion/ precipitation rates that cannot be easily obtained from conventional local mass transfer models and clearly illustrates the source of the differences between open pipe flows and closed loop flows.

V. Corrosion/Precipitation in Non-Isothermal and Multi-Modular LBE Loop Systems

Geometry changes in axis of liquid-metal coolant loop systems are unavoidable in engineering. On the other hand, corrosion and deposition of the materials themselves also change the flow area and then affect the corrosion/deposition process. To grasp the essences and mechanisms of the corrosion in a non-isotherm coolant loop system, the geometry variations must be considered. We extend the kinetic corrosion model for a simple loop to a model that can be used to calculate the corrosion/precipitation rate in a non-isothermal and multi-modular liquid-metal loop [J. Zhang and N. Li, *Journal of Nuclear Materials*, submitted 2003]. Several cases on the corrosion in LBE systems are studied to examine the combined effects of geometry variation and temperature profile. The model makes it possible to study the effects of corrosion and precipitation themselves on the corrosion and precipitation processes. The model is applied to an ideal loop with sinusoidal surface concentration profile and the DELTA loop to illustrate the combined effects of the axial conditions. It was found that:

1. The local corrosion/precipitation rate and the axial corrosion/precipitation profile depend on both the axial geometry variations and the axial temperature profile. The effects of the both axial conditions affect each other.
2. The local corrosion/precipitation rates increase with the increasing local wall shear rate. Depositions reduce the local flow area and result in a high local wall shear rate. The feedback of the deposition is positive and may quickly plug the loop structure, while the feedback of corrosion is negative.
3. Increasing or decreasing the flow area at one component may play an important role on corrosion at the other legs. The highest corrosion/precipitation does not necessarily occur at the leg where the velocity reaches its highest/lowest value; also it does not necessarily occur at the highest/lowest temperature. The locations of the highest corrosion and precipitation can be shifted by changing the hydraulic diameter at some leg and the axial temperature profile.
4. By only considering local condition effects, the local corrosion model may provide an incorrect value on the corrosion rate at the constant temperature leg in a non-isothermal and multi-modular loop system. One must consider the global operation conditions when analyzing experimental results from a test loop system.
5. The dependences of corrosion/precipitation on the hydraulic factors for a simple loop and a loop with multiple modules are significantly different. For the materials test loop, decreasing the tube diameter at the test leg results in a higher local corrosion rate and a smaller mass transfer coefficient.
6. The local mass transfer coefficient at the highest temperature leg decreases sharply at the beginning of the test leg and the change moves slowly downstream.

VI. Comparison Between Model Results and Experimental Data

Experimental results of corrosion tests performed in non-isothermal simple loop are available in pure liquid lead [J. Sannier and G. Santarini, *Journal of Nuclear Materials*, 107, 196 (1982)]. The authors found that the corrosion depth for steel 10 CD 9-10 was between 75-110 μm after 3000 hours and for Z 10 CD Nb V 92 steel is between 25 and 40 μm after 2800 hours. In our paper [J. Zhang and N. Li, *Journal of Nuclear Materials*, 321, 184 (2003)], we calculated the corrosion rate at the test section of the liquid-lead loop using the corrosion model for a simple loop. The model predicts an iron corrosion depth between 40 and 70 μm after 3000 hours, which agrees well with experimental results. The deviations are probably expected due to experimental uncertainties and alloy composition effects.

No experimental result on corrosion rate is available for LBE loops with oxygen control. An LBE loop called JLBL-1 loop [K. Kikuchi et al., *Journal of Nuclear Materials*, 318, 348 (2003)] was set up at the Japan Atomic Energy Research Institute, and some initial experiments have been carried out to study corrosion and deposition without actively controlling oxygen. All materials are stainless steel 316 (SS316). The inner diameters of the circulating tube, the test tube at the low temperature, and the test tube at high temperature are 22, 22, and 10 mm, respectively. The flow velocity is 1 m/s in the test tube and 0.2 m/s in other parts. The oxygen concentration is not measured during the experiment. The corrosion rate is less than 0.1 mm per 3000 hr at the test tube at the high temperature. Since there is no oxide layer reported and the experimental procedures seem to favor low initial oxygen concentration, we assume the corrosion process is mostly due to direct dissolution. To estimate corrosion/precipitation in the JLBL-1, the kinematic viscosity υ of LBE and the corrosion product diffusion coefficient D of the iron in LBE are needed. Both are functions of temperature. For the viscosity, we chose $\upsilon = 1.5 \times 10^{-7}\,\mathrm{m^2/s}$. For the diffusion coefficient, for which there are no directly experimental data, we chose $D_{\mathrm{Fe} \rightarrow \mathrm{Pb-Bi}} = 10^{-9}\,\mathrm{m^2/s}$. The calculated pure iron corrosion/precipitation profile for JLBL-1 shows that the deposition zones in the JLBL-1 loop can be exactly predicted using the present non-isothermal and multi-modular corrosion model. The predicted corrosion rate is about 0.025 mm per 3000 hr; that is close to the experimental value 0.03~0.1 mm. Taking into account that there are several uncertain factors (the value of the diffusion coefficient and the surface concentration, effects of the materials composition and erosion, etc.), the calculated values are consistent with the experiment results.

VII. TRAC Model

A TRAC model of the DELTA Loop has been developed [J. Lime and J. Zhang, *ACCAPP'03*, No-79245]. The modernized TRAC-M/F90 thermal-hydraulics code that is being developed by LANL for the United States Nuclear Regulatory Commission has been updated to include fluid properties used for advanced accelerator applications such as liquid sodium and LBE. The code also has the capability to track corrosion or oxidation products around the coolant loop. The TRAC DELTA Loop model has been benchmarked against a 48-hr steady-state test run that was performed August 6-8, 2002. TRAC-calculated temperatures were within the experimental uncertainty of the measured temperatures at the selected loop locations when external heat losses were accounted for. The TRAC model will be used for the pre-test prediction of steady-state and transient test runs, natural convection flow, corrosion, and safety analysis studies.

VII. Current Activities and Future Works

Active oxygen control in LBE can promote the formation of the "self-healing" oxide films on the structural material surface, drastically reducing steel corrosion and coolant contamination. By carefully controlling the amount of oxygen in LBE, it is possible to maintain an iron-and-chrome-oxide-based film on the structural steel surface. According to some experimental results, the protective oxide layers on steel structures are composed of an external layer (mainly Fe_3O_4) and an internal spinal layer. We are carrying out initial studies on the oxide growth in an LBE system. We have developed an oxide growth model for the pure metal case. We assumed that the oxide layer growth depth and the consumption depth of the metal obey the parabolic law and can be written as $X = 2\gamma_1(D_O^{ox}t)^{1/2}$, $Y = 2\gamma_2(D_O^{ox}t)^{1/2}$. We examined the parametric dependence of the growth constants γ_1 and γ_2. The reactions at the oxide/LBE and oxide/steel interface are considered. We plan to develop a model coupling the chemical reactions to the species diffusions through the oxide layer, the steel, and mass transport in hydrodynamics flows. Then we will provide a useful tool to interpret the test results in a quantitative framework and apply the data rationally for general applications. The ongoing experimental work in corrosion measurement will provide benchmarking data.

Finally, the present model assumes a mass transfer controlled corrosion process and neglects the transition effects. For high velocities, the surface reactions could control the corrosion processes. Other mechanisms such as erosion-corrosion or cavitation-corrosion need to be considered. In a practical LBE coolant system, there are many complex geometry structures, such as elbows, multi-branches, and gauges. Such geometry variations along the axis may result in high-intensity vortices and lead to a high local corrosion/precipitation rate. These effects will be considered in future modeling.

VIII. Publications List

1. Jinsuo Zhang and Ning Li, "Parametric Study of a Corrosion Model Applied to Lead-Bismuth Flow Systems," *Journal of Nuclear Materials*, 321,184 (2003).
2. Jinsuo Zhang and Ning Li, "Improved Application of Local Models to Steel Corrosion in Lead-Bismuth Loops," *Nuclear Technology*, 144 (12), NT-002115 (2003).
3. Jinsuo Zhang and Ning Li, "A Correlation of Steel Corrosion in Non-isothermal LBE Loop Systems," *Journal of Nuclear Science and Technology*, (accepted, will appear 2003).
4. Jinsuo Zhang and Ning Li, "Analytical Solution on Transient Corrosion/ Precipitation in Closed Loop Systems," *Corrosion*, (accepted, will appear 2003).
5. Jinsuo Zhang and Ning Li, "Corrosion/Precipitation in Non-Isothermal and Multi-Modular LBE Loop," *Journal of Nuclear Materials*, (submitted, will appear 2003).
6. Jinsuo Zhang and Ning Li, "A Kinetic Model on Corrosion/precipitation in Lead-Bismuth Eutectic Loop," *Proceeding of the Sixth International Meeting on Nuclear Applications of Accelerator Technology (AccApp'03)*, No-79709.
7. James F. Lime and Jinsuo Zhang, "A TRAC Model of the Los Alamos National Laboratory DELTA Loop Facility," *Proceeding of the Sixth International Meeting on Nuclear Applications of Accelerator Technology (AccApp'03).* NO-79245.
8. C. Wu, K. Dasika, Y. Chen, S. Moujaes, J. Zhang, and N. Li, "Study of Geometry Effects on Local Corrosion Rates for LBE Loop," *The ANS National Conference San Diego June 2003* (accepted, will appear 2003).
9. K. Dasika, C. Wu, S. Moujaes, Y. Chen, N. Li, and J. Zhang, "Simulation Considerations in Lead-Bismuth Transmutation Loops: Corrosion Concentration, Velocity and Temperature Profiles of LBE Loops," *NURETH10-conf*. S. Korea Oct. 5-9, 2003.
10. Ning Li and Jinsuo Zhang, "Modeling Corrosion in Oxygen Controlled LBE Systems," *The 11th International Conference on Nuclear Engineering*, ICONE11-36560.